ESRF	**Experiment title:** **Pressure and magnetic field studies of superconducting $Li_x(NH_2)_y(NH_3)_{1-y}Fe_2Se_2$ using ^{57}Fe Synchrotron Mössbauer Source**	**Experiment number**: HC-1668
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In the beamtime HC-1668, we investigated an NH_3/Li intercalate of FeSe exhibiting superconductivity below 42 K. This novel high-T_C superconductor was synthesized by Burrard-Lucas *et al.* [1] and characterized by the applicants using Mössbauer spectroscopy at ambient pressure [2]. It was shown that superconducting transition in the intercalated FeSe is accompanied by appearance of an additional magnetic spectrum implying strong magnetic fluctuations (see Fig. 2 in Application for HC-1668). These fluctuations are believed to play a major role in the pairing mechanism of superconducting charge carries of Fe-based superconductors [3, 4]. In this regard, the NH_3/Li intercalate of FeSe with its high T_C could be a clue compound for the understanding of the principal mechanisms of superconductivity in these systems. Taking into account our recent studies of conductivity in intercalated FeSe showing a strong decrease of T_C with pressure (Fig. 3 in Application), a detailed Mössbauer study of this compound under pressure was performed using the Synchrotron Mössbauer Source (SMS) at ID18.

We used ^{57}Fe-SMS to study the hyperfine parameters of the NH_3/Li intercalated FeSe as function of temperature (3 K, 10 K, 20 K, 40 K, 300 K) as well as pressure (ambient, 2.1 GPa, 3.2 GPa, 5.3 GPa, 13.3 GPa). Fig. 1 shows selected ^{57}Fe-spectra of NH_3/Li intercalated FeSe at 3 K at various pressures. At ambient pressure, the spectrum consists of a dominant quadrupole doublet with values for the isomer shift δ and quadrupole splitting ΔE_Q very near the ones in [2]. The magnetic subspectrum, visible only in the enlarged velocity range, reflects strong magnetic fluctuations. Temperature dependent measurements on the sample at ambient pressure reveal increasing of the magnetic fluctuations frequency and a continuous decrease of the magnetic site intensity at high temperatures. This behaviour follows closely the superconducting transition curve as monitored by magnetic susceptibility. Therefore we consider these magnetic fluctuations to be strongly associated with superconductivity.

Next we studied with ^{57}Fe-SMS the effect of pressure on the hyperfine parameters in NH_3/Li intercalated FeSe for comparison with the pressure dependence of T_C. These studies delivered three main results:

1) Upon increasing pressure, the isomer shift of the main non-magnetic site dramatically decreases (Fig. 2). This observation is due to an increasing of *s*-electron density on Fe nuclei. Apparently, redistribution of electron density may affect the density of states at Fermi level resulting in changes of superconducting properties. According to our conductivity measurements on the intercalated FeSe, T_C goes down with pressure. Therefore the change of δ with pressure can be attributed to a pressure-induced redistribution of *s*- and *d*-electron density in the conduction band and agrees well with behaviour of T_C [2].

2) The quadrupole splitting ΔE_Q of the main doublet decreases with pressure, and at 5.3 and 13.3 GPa the spectra can be fitted with the single line only. Such behaviour of ΔE_Q is unusual for the present systems, because ΔE_Q is expected to increase with compression of the local surrounding. This behaviour may point to a structural phase transition, starting at 5 GPa, as indicated by the strongly discontinuous variation of the isomer shift as well (Fig. 2). To elucidate this effect, structural studies at pressures above 5 GPa and also corresponding band structure calculations for δ and ΔE_Q are needed.

3) The magnetic subspectra reflect an increase of the magnetic fluctuation frequency, as reflected by the broadened shape of the subspectrum at 2.1 GPa (Fig. 1). This increase of magnetic fluctuation frequency is connected with a decrease of the relative amount of the magnetic part, from 26% at ambient pressure quite slowly to 24% at 3.2 GPa, then rapidly and being completely absent at 13.3 GPa.

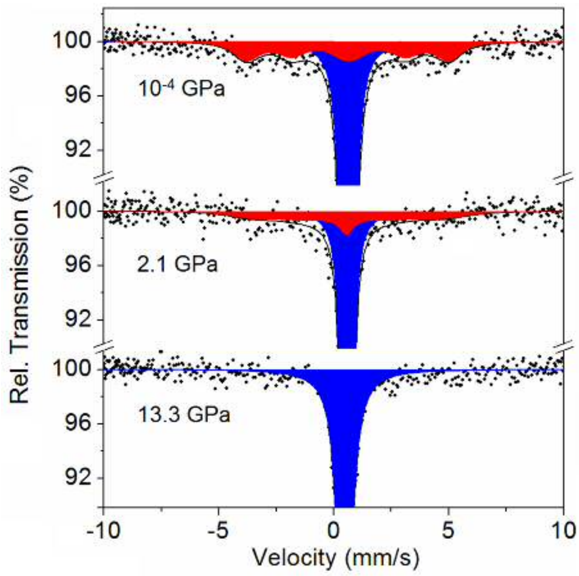

Fig. 1: Selected Mössbauer spectra of intercalated FeSe at ambient pressure, 2.1 and 13.3 GPa (T = 3 K), showing behaviour of the magnetic subspectra with pressure.

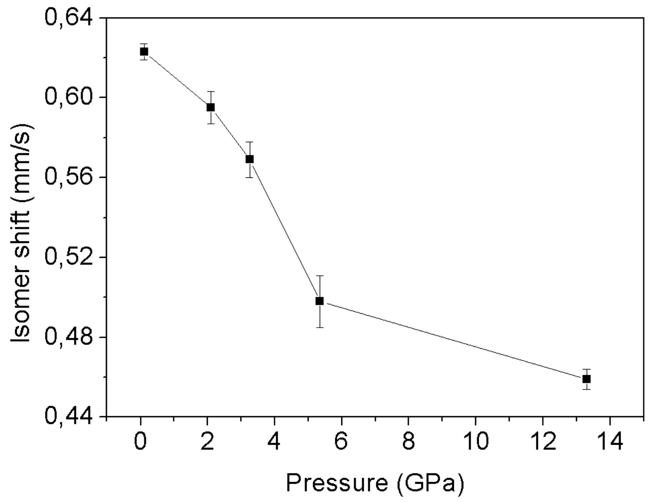

Fig. 2: Pressure dependence of the isomer shift for the intercalated FeSe, indicating a possible pressure-induced phase transition above 5 GPa (T = 3 K).

The present observations of the magnetic subspectra in NH_3/Li intercalated FeSe support our assumption that magnetic fluctuations are relevant for the pairing mechanism in FeSe-based superconductors. The behaviour of the magnetic fraction in the spectra at ambient pressure strongly correlates with the superconducting transition. Both the amount of magnetic fraction and the frequency of its fluctuations do follow the variation of T_C with pressure.

During the HC-1668 beamtime, we performed preliminary ^{57}Fe-SMS pressure studies of Cu-doped FeSe superconductor (20% ^{57}Fe-enriched $Cu_{0.04}Fe_{0.97}Se$). In contrast with the NH_3/Li intercalated sample, the isomer shift of Cu-doped FeSe increases upon increasing pressure, while the amount of the magnetic subspectrum decreases. Most interestingly, the T_C values of $Cu_{0.04}Fe_{0.97}Se$ increases with pressure [5]. These preliminary results point to a correlation between the isomer shift and T_C in FeSe-based superconductors as well. Detailed investigation of Cu-doped FeSe is envisaged in our following ESRF proposal.

References

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